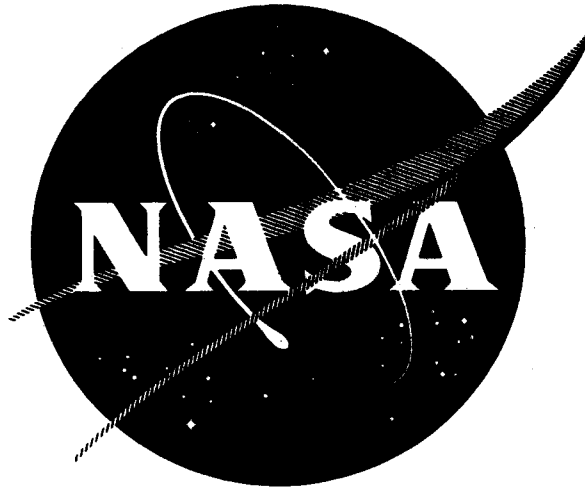


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**CALCULATION OF SLOW NEUTRON SCATTERING CROSS-SECTIONS OF MOLECULAR
HYDROGEN AND DEUTERIUM**

by

James A. Young and Juan U. Koppel

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TOPICAL REPORT

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ABSTRACT

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A

The cross-sections for neutrons scattering from H_2 and D_2 has been calculated taking into account the spin correlations, rotations, and vibrations of the molecules exactly, to the extent that the vibrations are harmonic and do not interact with the rotations. Free translations of the molecules are assumed, but this assumption is expected to be valid in the liquid for neutron energies above .002 ev. Numerical results are given for the total cross-section for both ortho- and para-hydrogen gas at $20.4^\circ K$ which agree reasonably well with the limited experimental results available. Also curves of the double differential cross-section are shown for selected incident neutron energies and scattering angles. These latter curves show very clearly the various rotational and vibrational transitions. The formulae given here are applicable at all temperatures below the thermal excitation of the first vibrational level.

A. A. A.

I. INTRODUCTION

In recent years, liquid hydrogen has become important as a propellant in nuclear rockets. This application necessitates a knowledge of the interactions of neutrons with the H_2 molecule. Secondary γ -heating due to absorption of neutrons by hydrogen atoms must be predicted, and the neutron shielding properties of liquid hydrogen must be understood. For both of these applications it is necessary to know how the neutrons are transported through the liquid and thus the cross-section and distribution of neutron scatterings from H_2 molecules must be predicted.

Several Monte Carlo computer codes have been written for use in solving propellant heating and shielding problems. Each of these codes uses a particular set of assumptions in the slow neutron energy range. For example, one code currently in use assumes isotropic scattering from free protons for neutron energies above .08 ev. The results in this paper show that while this assumption is not too severe for the total cross-section, it is far from true for the angular distribution of scattered neutrons. In addition, vibrations of the H_2 molecule are completely ignored in the code, but are included exactly in this work.

The technique of slow neutron scattering has proved most valuable as a probe for studying molecular systems,⁽¹⁾ and much work has been devoted to the development of calculational methods and models which will adequately describe the scattering from such systems. The H_2 molecule is not only perhaps the simplest of all but, as a liquid, hydrogen is important both as a cold neutron source and, in special cases, as a shield. However, up to the present, the only calculations that have been performed^(2,3) are applicable

to high temperatures ($\approx 400^\circ\text{K}$) with high incident neutron energies ($\geq .06$ ev), and to low temperatures ($\leq 30^\circ\text{K}$) with low neutron energies ($\leq .05$ ev).^(4,5)

The purpose of this present paper is to derive the cross-section for neutrons scattering from hydrogen gas in the energy range from 0 ev to roughly 3 ev, and for temperatures $\leq 3000^\circ\text{K}$ (the vibrational levels are assumed unpopulated). The results are applicable to liquid hydrogen, with the restriction that for incident neutron energies less than that of thermal equilibrium ($\leq .002$ ev) the results are not expected to be accurate. Spin correlations, rotations and vibrations are taken into account exactly, to the extent that vibration-rotation coupling can be neglected, and that the vibrations are harmonic. Some numerical results are given, and these are compared with available experimental data. It is hoped that when more detailed experimental information is obtained, the calculation given here will prove useful in evaluating the assumptions that enter into the theory for more complicated molecules.

Since the only significant calculational difference between D_2 and H_2 is the effect of spin correlations, these have been evaluated, and thus we have derived the cross-section for D_2 in addition to that for H_2 .

II. GENERAL FORMULATION

In order to describe the interaction of a neutron with a molecule, the specifically nuclear interaction is replaced by a point delta-function interaction, with the amplitude adjusted to give the correct cross-section for scattering from isolated and fixed nuclei. With this provision the first Born approximation can be used to describe the scattering of neutrons with the complete molecular system. If the position vector of a nucleus is \vec{r} and that of the neutron is \vec{r}_n , then the potential experienced by the neutron is

$$V(\vec{r}_n, \vec{r}) = (2\pi a/m) \delta(\vec{r}_n - \vec{r})$$

where m is the neutron mass and a the amplitude, or in other words the bound scattering length, which is in general spin-dependent.

If now one uses the first Born approximation:

$$\frac{d^2\sigma}{d\Omega d\epsilon} = \left(\frac{m}{2\pi}\right)^2 \frac{k}{k_0} \left| \langle \psi_f | \int d^3r_n e^{-i(\vec{k}-\vec{k}_0)\cdot\vec{r}_n} V(\vec{r}_n) | \psi_i \rangle \right|^2,$$

and the representation:

$$2\pi \delta(x) = \int_{-\infty}^{\infty} dt e^{ixt},$$

of the delta-function, which serves to express the energy conservation condition in time-dependent form, the cross-section for neutrons scattering from nuclei bound in any chemical system can be found. The problem is then reduced to a consideration of the dynamics of the scatterer.

In units where $\hbar = 1$, the differential cross-section for the scattering of neutrons from any molecular system with initial wave function ψ_i and final wave function ψ_f is⁽¹⁾

$$\frac{d^2\sigma}{d\Omega d\epsilon} = \frac{1}{2\pi} \frac{k}{k_0} \sum_f \int_{-\infty}^{\infty} dt e^{+i\epsilon t} \left[e^{-i(E_i - E_f)t} \left| \langle \psi_f | \sum_m e^{i\vec{k} \cdot \vec{r}_m} a_m | \psi_i \rangle \right|^2 \right]_T. \quad (1)$$

Here k_0 , k are initial and final neutron momenta respectively, E_i , E_f are the initial and final neutron energies, ϵ the neutron energy transfer, \vec{k} the neutron momentum transfer, \vec{r}_m and a_m the position vector and scattering amplitude of the m -th atom respectively, and the subscript T implies that a thermal average is to be taken over the initial states.

The H_2 molecule is dumbbell-shaped with a separation a . The molecule can then vibrate along the line joining the atoms, and rotate about the center-point of that line. We write then for the position vector of a hydrogen atom:

$$\vec{r}_m = \vec{r}_l + (-)^n \vec{R}_l / 2, \quad n = 1, 2$$

where \vec{r}_l is the position coordinate of the molecular center-of-mass, and \vec{R}_l the relative coordinate of the two atoms of the l -th molecule. Using this separation Eq. (1) becomes

$$\begin{aligned}
\frac{d^2\sigma}{d\Omega d\epsilon} = & \frac{1}{2\pi} \frac{k}{k_0} \sum_f \int_{-\infty}^{\infty} dt \left[e^{+i(\epsilon - E_1 + E_f)t} \sum_{\lambda \neq j} \sum_{n,p=1}^2 \langle \psi_1 | a_{jp} e^{-i\vec{k} \cdot \vec{r}_j - i(-)^{p \rightarrow} \vec{R}_j \cdot \vec{\kappa}/2} | \psi_f \rangle \right. \\
& \left. \cdot \langle \psi_f | a_{\lambda n} e^{i\vec{k} \cdot \vec{r}_\lambda + i(-)^{n \rightarrow} \vec{R}_\lambda \cdot \vec{\kappa}/2} | \psi_1 \rangle \right]_T \\
& + \frac{1}{2\pi} \frac{k}{k_0} \sum_f \int_{-\infty}^{\infty} dt \left[e^{+i(\epsilon - E_1 + E_f)t} \sum_{\lambda} \sum_{n,p=1}^2 \langle \psi_1 | a_{\lambda p} e^{-i\vec{k} \cdot \vec{r}_\lambda - i(-)^{p \rightarrow} \vec{\kappa} \cdot \vec{R}_\lambda/2} | \psi_f \rangle \right. \\
& \left. \cdot \langle \psi_f | a_{\lambda n} e^{i\vec{k} \cdot \vec{r}_\lambda + i(-)^{n \rightarrow} \vec{\kappa} \cdot \vec{R}_\lambda/2} | \psi_1 \rangle \right]_T \quad (2)
\end{aligned}$$

First we consider the second term which refers to a single H_2 molecule, and which we refer to as the "self" term. If we make the good assumption that the translational modes can be separated, then we can write for the "self" term

$$\begin{aligned}
\frac{d^2\sigma_s}{d\Omega d\epsilon} = & \frac{1}{2\pi} \frac{k}{k_0} \sum_f \int_{-\infty}^{\infty} dt \sum_{\lambda} \langle \psi_{1t} | e^{-i\vec{k} \cdot \vec{r}_\lambda(0)} e^{i\vec{k} \cdot \vec{r}_\lambda(t)} | \psi_{1t} \rangle_T \\
& \left[\cdot \left| \langle \psi_f | \sum_{n=1}^2 a_{\lambda n} e^{(-)^{n \rightarrow} i\vec{k} \cdot \vec{R}_\lambda/2} | \psi_1 \rangle \right|^2 e^{+i(\epsilon - E_1' + E_f')t} \right]_T \quad (3)
\end{aligned}$$

where ψ_{it} is the initial wave function of the translational modes, and E'_i, E'_f are the energies of the rotational and vibrational states only. It is well to point out here that due to the vibrations R is not constant.

2.1 Spin Correlations

The nuclei of the H_2 molecule can form states of total spin $S=0$ or $S=1$, the former corresponding to para- and the latter to ortho-hydrogen. For $S=0$ only states of total angular momentum J having even values can occur, and for $S=1$ only odd values of J are allowed. In this section we compute the effects in Eq. (3) that are due to the nuclear spins. Call

$$g(t) = \left[\sum_f \left| \langle \psi_f | \sum_{n=1}^2 a_n e^{(-) i \kappa \cdot R/2} | \psi_i \rangle \right|^2 e^{-i(E'_i - E'_f)t} \right]_T. \quad (4)$$

We can write $g(t)$ explicitly in the form

$$g(t) = \sum_{J,S} \frac{P_{JS}}{2J+1} \frac{1}{2S+1} \frac{1}{2} \sum_{\substack{\sigma_z = \pm 1/2 \\ \sigma'_z = \pm 1/2}} \sum_{J'} e^{i(E_{J'} - E_J)t} \sum_{n=0} e^{inmt}.$$

$$\sum_{J_z J'_z} \sum_{S'} \sum_{S'_z} \left| \langle J'_z J'_z, S'_z S'_z, \sigma'_z, n | A | J J_z, S S_z, \sigma_z, n=0 \rangle \right|^2, \quad (5)$$

which includes a thermal average over the initial states,

where the parameters are defined as follows:

J, J'	initial and final angular momentum of the molecule (J_z and J'_z are their z component)
S, S'	initial and final total spin of the molecule (S_z and S'_z are their z component)
σ_z, σ'_z	initial and final z component of the neutron spin
E_J	rotational energy of the state J
P_{JS}	statistical weight of the state J with spin S
ω	quantum of vibrational energy
n	vibrational quantum number

$$A = a_1 e^{i\kappa \cdot \frac{\vec{R}}{2}} + a_2 e^{-i\kappa \cdot \frac{\vec{R}}{2}}$$

Thus $g(t)$ refers to the rotational, vibrational and spin dependent part of Eq. (3). The molecular Hamiltonian has been taken to be spin independent and as stated previously the coupling between rotations and vibrations is neglected. Thus $|J, S, n\rangle = |J\rangle |S\rangle |n\rangle$.

Since $\hbar\omega = .546$ ev, for hydrogen all the molecules are initially in their vibrational ground state, $n=0$, the higher states being frozen out except at very high temperature ($\gtrsim 3000^\circ\text{K}$). The rotational energy levels are given by

$$E_J = \frac{\hbar^2 J(J+1)}{4Ma^2} = 0.015 \frac{J(J+1)}{2} \text{ ev}, \quad (6)$$

where M is the proton mass, and a the equilibrium separation distance of the H-H bond. Now let us rewrite the operator A . We recall that the scattering length operator of the proton has the form

$$a_m = \frac{I_m + 1 + 2 \vec{I}_m \cdot \vec{\sigma}}{2I_m + 1} a_+ + \frac{I_m - 2\vec{I}_m \cdot \vec{\sigma}}{2I_m + 1} a_- \quad (7)$$

where I_m is the spin of the proton and a_+ and a_- are the triplet and singlet scattering lengths, respectively of the neutron-proton interaction. Since $I = 1/2$ and $\vec{I}_1 + \vec{I}_2 = \vec{S}$ we find

$$\begin{aligned} \frac{A}{2} = a_{\text{coh}} \cos \frac{\vec{\kappa} \cdot \vec{R}}{2} + \frac{2}{\sqrt{3}} a_{\text{inc}} \left[\cos \left(\frac{\vec{\kappa} \cdot \vec{R}}{2} \right) \vec{\sigma} \cdot \vec{S} + \right. \\ \left. + i \sin \left(\frac{\vec{\kappa} \cdot \vec{R}}{2} \right) \vec{\sigma} \cdot (\vec{I}_1 - \vec{I}_2) \right] \end{aligned} \quad (8)$$

with the usual definitions of coherent and incoherent scattering length:

$$\begin{aligned} a_{\text{coh}}^2 = \langle a \rangle^2 = \frac{1}{(2I+1)^2} \left[(I+1)a_+ + I a_- \right]^2 = \\ = \frac{1}{16} (3 a_+ + a_-)^2, \end{aligned} \quad (9)$$

$$\begin{aligned} a_{\text{inc}}^2 = \langle a^2 \rangle - \langle a \rangle^2 = \frac{I(I+1)}{(2I+1)^2} (a_+ - a_-)^2 = \\ = \frac{3}{16} (a_+ - a_-)^2, \end{aligned} \quad (10)$$

respectively.

First we shall consider transitions between states of the same parity (and hence with the same molecular spin S). In this case only the symmetric part of A contributes to the matrix elements and calling α and β the initial and final states we have

$$|\langle \beta | A | \alpha \rangle_{S=S}|^2 = |\langle J' J'_n | \cos(\frac{\vec{\kappa} \cdot \vec{R}}{2}) | J J_n n=0 \rangle|^2 |\langle \sigma'_z S' S'_z | P | \sigma_z S S_z \rangle|^2 \quad (11)$$

where

$$P = 2 a_{\text{coh}} + \frac{4}{\sqrt{3}} a_{\text{inc}} \vec{\sigma} \cdot \vec{S}$$

Now we may sum over the final spin states with the result

$$\sum_{\sigma'_z S' S'_z} |\langle \beta | A | \alpha \rangle|^2 = |\langle J' J'_n | \cos(\frac{\vec{\kappa} \cdot \vec{R}}{2}) | J J_n n=0 \rangle|^2 \langle \sigma_z S S_z | P^2 | \sigma_z S S_z \rangle \quad (12)$$

The square of the hermitian operator P is

$$P^2 = P^\dagger P = 4a_{\text{coh}}^2 + \frac{16}{3} a_{\text{inc}}^2 (\vec{\sigma} \cdot \vec{S})^2 + \frac{16}{\sqrt{3}} a_{\text{coh}} a_{\text{inc}} \vec{\sigma} \cdot \vec{S} \quad (13)$$

Summing over the neutron spin states the last term averages to zero. Since

$$\sum_{\sigma_z} \langle \sigma_z | (\vec{\sigma} \cdot \vec{I}_\nu) (\vec{\sigma} \cdot \vec{I}_\mu) | \sigma_z \rangle = \frac{1}{2} \vec{I}_\nu \cdot \vec{I}_\mu \quad (14)$$

or

$$\sum_{\sigma_z} \langle \sigma_z | (\vec{\sigma} \cdot \vec{S})^2 | \sigma_z \rangle = \frac{1}{2} S^2$$

we get

$$\sum_{\sigma_z} \langle \sigma_z S S_z | P^2 | \sigma_z S S_z \rangle = 8 \left[a_{\text{coh}}^2 + \frac{1}{3} a_{\text{inc}}^2 S(S+1) \right] \quad (15)$$

Substituting now Eq. (12) and (15) into (5) and summing over S , we obtain for spin conserving transitions

$$g_{S=S'}(t) = 4 \sum_J \frac{P_J}{2J+1} \left[a_{\text{coh}}^2 + \frac{a_{\text{inc}}^2}{3} S(S+1) \right] \sum_{J'} e^{i(E_{J'} - E_J)t} \quad (16)$$

$$\cdot \sum_{n=0} e^{in\omega t} \sum_{J_z J'_z} \left| \langle J' J'_z n | \cos\left(\frac{\vec{\kappa} \cdot \vec{R}}{2}\right) | J J_z n=0 \rangle \right|^2$$

where the sum $\sum_{J'}$ is over states of same parity as J .

For transitions between states of opposite parity (and hence of different spin S) only the antisymmetric part of the operator A gives a non-vanishing contribution to the matrix elements. In this case

$$\left| \langle \beta | A | \alpha \rangle \right|_{S \neq S'}^2 = \left| \langle J' J'_z n | \sin\left(\frac{\vec{\kappa} \cdot \vec{R}}{2}\right) | J J_z n=0 \rangle \right|^2 \left| \langle \sigma'_z S' S'_z | Q | \sigma_z S S_z \rangle \right|^2 \quad (17)$$

with

$$Q = \frac{4}{\sqrt{3}} a_{\text{inc}} \vec{\sigma} \cdot (\vec{I}_1 - \vec{I}_2) \quad (18)$$

Again, summing over final spin states and considering that

$$\sum_{\sigma_z} \langle \sigma_z | \left[\vec{\sigma} \cdot (\vec{I}_1 - \vec{I}_2) \right]^2 | \sigma_z \rangle = \frac{1}{2} (\vec{I}_1 - \vec{I}_2)^2 = \vec{I}_1^2 + \vec{I}_2^2 - \frac{1}{2} \vec{S}^2, \quad (19)$$

one finds

$$g_{S \neq S'}^{(t)} = 4 \sum_J \frac{P_J}{2J+1} a_{inc}^2 \left[1 - \frac{S(S+1)}{3} \right] \sum_{J'} e^{i(E_{J'} - E_J)t} \cdot \sum_{n=0} e^{in\omega t} \sum_{J_z J_z'} \left| \langle J' J_z' n | \sin \frac{\vec{\kappa} \cdot \vec{R}}{2} | J J_z n=0 \rangle \right|^2 \quad (20)$$

where the sum $\sum_{J'}$ is over states of opposite parity to J . These same results can be obtained for D_2 and are given in Appendix A.

2.2 Vibrations

Since all the molecules are initially in their ground vibrational state, the vibrational transitions are important only if the incident neutron energy is great enough so that $\kappa^2/8M\omega \geq 1$. The neutron then cannot gain energy from the vibrational modes.

The vibrational matrix elements can be evaluated exactly under the assumption that the restoring force of the H-H bond is harmonic in nature. We write $R = a+x$ where x is the amount the bond length is stretched. On expanding x ,

$$x = i(M\omega)^{-1/2} [b - b^+]$$

where b^+ , b are the boson creation and annihilation operators respectively, the matrix elements in Eqs. (16) and (20) are of the form

$$\langle n | e^{\pm \frac{i\kappa R \mu}{2}} | 0 \rangle = e^{\pm \frac{i\kappa a \mu}{2}} \langle n | e^{\mp \frac{\kappa \mu}{2\sqrt{M\omega}} (b - b^+)} | 0 \rangle$$

where $\mu = \cos \theta$, and θ is the angle between $\vec{\kappa}$ and \vec{R} .

Using the relation

$$e^A e^B = e^{A+B+1/2 [A,B]} ,$$

the above expression becomes:

$$\begin{aligned} & \langle n | e^{\pm \frac{i \kappa R \mu}{2}} | 0 \rangle \\ &= \exp \left(\pm \frac{i \kappa a \mu}{2} \right) \exp \left(-\frac{\kappa^2 \mu^2}{8 M \omega} \right) \langle n | \exp \left(\pm \frac{\kappa \mu b^+}{2 \sqrt{M \omega}} \right) \exp \left(\mp \frac{\kappa \mu b}{2 \sqrt{M \omega}} \right) | 0 \rangle \\ &= \left(\frac{\pm \kappa \mu}{2 \sqrt{M \omega}} \right)^n \frac{1}{\sqrt{n!}} \exp \left(\pm \frac{i \kappa a \mu}{2} \right) \exp \left(-\frac{\kappa^2 \mu^2}{8 M \omega} \right) . \end{aligned}$$

One readily observes then that in Eq. (16)

$$\begin{aligned} & \left| \langle J' J_z' n | \cos \left(\frac{\vec{\kappa} \cdot \vec{R}}{2} \right) | J J_z n=0 \rangle \right|^2 \\ &= \left(\frac{\kappa^2}{4 M \omega} \right)^n \frac{1}{n!} \left| \langle J' J_z' | \mu^n \exp \left(-\frac{\kappa^2 \mu^2}{8 M \omega} \right) \left(e^{\frac{i \kappa a \mu}{2}} + (-)^n e^{-\frac{i \kappa a \mu}{2}} \right) | J J_z \rangle \right|^2 \\ &= \left(\frac{\kappa^2}{4 M \omega} \right)^n \frac{1}{n!} \left| \langle J' J_z' | \mu^n \exp \left(-\frac{\kappa^2 \mu^2}{8 M \omega} \right) e^{\frac{i \kappa a \mu}{2}} | J J_z \rangle \right|^2 , \end{aligned} \quad (21)$$

since J' and J have the same parity. A similar expression holds if J' and J have opposite parity, so that Eqs. (16) and (20) become

$$\begin{aligned} g(t)_{S=S'} &= 4 \left(a_{\text{coh}}^2 + \frac{a_{\text{inc}}^2}{3} S(S+1) \right) \sum_J \frac{P_{JS}}{2J+1} \sum_{J'} e^{i(E_{J'} - E_J)t} . \\ & \cdot \sum_n e^{i n \omega t} \left(\frac{\kappa^2}{4 M \omega} \right)^n \frac{1}{n!} \sum_{J_z J_z'} \left| \langle J' J_z' | \mu^n \exp \left(-\frac{\kappa^2 \mu^2}{8 M \omega} + \frac{i \kappa a \mu}{2} \right) | J J_z \rangle \right|^2 \end{aligned} \quad (22)$$

$$g(t)_{S \leftrightarrow S} = 4 a_{inc}^2 \left(1 - \frac{S(S+1)}{3} \right) \sum_J \frac{P_{JS}}{2J+1} \sum_{J'} e^{i(E_{J'} - E_J)t} \quad (23)$$

$$\cdot \sum_{n=0} e^{in\omega t} \left(\frac{\kappa^2}{4M\omega} \right)^n \frac{1}{n!} \sum_{J_z J_z'} \left| \langle J' J_z' | \mu^n \exp \left(-\frac{\kappa^2 \mu^2}{8M\omega} + \frac{i\kappa a \mu}{2} \right) | J J_z \rangle \right|^2$$

Eqs. (22) and (23) are expected to be valid for n up to a value of 5, for above this value the H-H bond must begin to show its non-linear character, and the harmonic approximation should fail.

2.3 Rotations

Since $E_J = \frac{.015}{2} J(J+1)$, at liquid hydrogen temperature, $kT \ll .015$ and all the molecules are in their lowest rotational state, which is $J=0$ for para-hydrogen and $J=1$ for ortho-hydrogen. At higher temperatures however, we must know the transition probability between two arbitrary rotational states.

The wave functions for the rotational states of the linear H_2 molecule (if we assume no rotational-vibration coupling, and that there is no hindrance to the rotation) are just the spherical harmonics:

$$|J J_z\rangle = Y_{J J_z}(\theta, \phi).$$

We choose a coordinate system with $\vec{\kappa}$ along the z -axis. The rotational matrix elements then are of the form:

$$\begin{aligned} \langle J' J_z' | \mu^n \exp \left(-\frac{\kappa^2 \mu^2}{8M\omega} + i\frac{\kappa a \mu}{2} \right) | J J_z \rangle = \\ = \int d\Omega \mu^n \exp \left(-\frac{\kappa^2 \mu^2}{8M\omega} + i\frac{\kappa a \mu}{2} \right) Y_{J' J_z'}^*(\theta, \phi) Y_{J J_z}(\theta, \phi) \end{aligned} \quad (24)$$

If now we make use of the coupling theorem⁽⁶⁾ for the spherical harmonics, namely:

$$Y_{l_1 m_1}(\theta, \phi) Y_{l_2 m_2}(\theta, \phi) = \sum_{l=|l_1-l_2|}^{l_1+l_2} \left[\frac{(2l_1+1)(2l_2+1)}{4\pi(2l+1)} \right]^{1/2} C(l_1 l_2 l; m_1 m_2) C(l_1 l_2 l; 00) Y_{l, m_1+m_2}(\theta, \phi),$$

where the $C(l_1 l_2 l; m_1 m_2)$ are the Clebsch-Gordan coefficients which vanish (for $m_1=m_2=0$) unless $l_1 + l_2 + l$ is even, and notice that J'_z must equal J_z , Eq. (24) can be written as:

$$\begin{aligned} & \langle J' J'_z | \mu^n \exp \left(-\frac{\kappa^2 \mu^2}{8M\omega} + i \frac{\kappa a \mu}{2} \right) | J J_z \rangle \\ &= \delta_{J'_z J_z} (-)^{J'_z} \sum_{l=|J'-J|}^{J'+J} \left[\frac{(2J'+1)(2J+1)}{4\pi(2l+1)} \right]^{1/2} C(J J' l; J_z - J'_z) C(J J' l; 00) \cdot \\ & \quad \cdot \int d\Omega \mu^n \exp \left(-\frac{\kappa^2 \mu^2}{8M\omega} + i \frac{\kappa a \mu}{2} \right) Y_{l, J_z - J'_z}(\theta, \phi), \end{aligned}$$

thus

$$\begin{aligned} \sum_{J'_z J_z} | \langle J' J'_z | \mu^n \exp \left(-\frac{\kappa^2 \mu^2}{8M\omega} + i \frac{\kappa a \mu}{2} \right) | J J_z \rangle |^2 &= \frac{(2J'+1)(2J+1)}{4} \cdot \\ & \cdot \sum_{l=|J'-J|}^{J'+J} |A_{nl}|^2 C^2(J J' l; 00) \end{aligned} \quad (25)$$

where we have used the closure relation for the Clebsch-Gordan coefficients:

$$\sum_{J_z} C(J J' \ell'; J_z - J_z) C(J J' \ell; J_z - J_z) = \delta_{\ell \ell'}.$$

The $A_{n\ell}$ are defined by

$$A_{n\ell} = \int_{-1}^1 d\mu \mu^n \exp \left(-\frac{\kappa^2 \mu^2}{8M\omega} + \frac{i\kappa a \mu}{2} \right) P_\ell(\mu),$$

and $P_\ell(\mu)$ is the Legendre polynomial of order ℓ .

Using Eq. (25), Eqs. (22) and (23) become

$$g_{S=S'}(t) = \left[a_{\text{coh}}^2 + \frac{a_{\text{inc}}^2}{3} S(S+1) \right] \sum_J P_J \sum_{J'} e^{i(E_{J'} - E_J)t} (2J'+1) \cdot$$

$$\cdot \sum_{n=0} e^{in\omega t} \left(\frac{\kappa^2}{4M\omega} \right)^n \frac{1}{n!} \sum_{\ell=|J'-J|}^{J'+J} |A_{n\ell}|^2 C^2(J J' \ell; 00) \quad (26)$$

$$g_{S \neq S'}(t) = a_{\text{inc}}^2 \left(1 - \frac{S(S+1)}{3} \right) \sum_J P_J \sum_{J'} e^{i(E_{J'} - E_J)t} (2J'+1) \cdot$$

$$\cdot \sum_{n=0} e^{in\omega t} \left(\frac{\kappa^2}{4M\omega} \right)^n \frac{1}{n!} \sum_{\ell=|J'-J|}^{J'+J} |A_{n\ell}|^2 C^2(J J' \ell; 00) \quad (27)$$

The coefficients $C^2(J J' \ell; 00)$ are given in Appendix B for $J = 0, 1, 2, 3, 4$.

2.4 Translations

For hydrogen we assume that the molecular translations are free. Using familiar results,⁽⁵⁾ the translational part of Eq. 3 becomes

$$\langle \psi_{it} | e^{-i\vec{\kappa} \cdot \vec{r}_l(0)} e^{i\vec{\kappa} \cdot \vec{r}_l(t)} | \psi_{it} \rangle_T = e^{-\frac{t\kappa^2}{4M}(1+tT)}, \quad (28)$$

where the temperature T is measured in electron-volts.

The above approximation is likely to be a very good one for gaseous hydrogen, but for neutron scattering in liquid hydrogen, this approximation will generally be invalid if the initial neutron energy is below the liquid hydrogen thermal energy.

III. SCATTERING CROSS-SECTIONS

By using Eqs. (26), (27) and (28) in Eq. (3), and performing the indicated integration over t , the "self" scattering cross-section per molecule from para-hydrogen becomes:

$$\begin{aligned}
 \left. \frac{d^2 \sigma_s}{d\Omega d\epsilon} \right|_{\text{para}} &= \frac{k}{k_0} \sqrt{\frac{M}{\pi \kappa^2 T}} \sum_n \frac{1}{n!} \left(\frac{\kappa^2}{4M\omega} \right)^n \sum_{J=0,2,4,\dots} P_J \left[a_c^2 \sum_{J'=0,2,4,\dots} \right. \\
 &\cdot (2J'+1) \exp \left\{ - \left(\epsilon + \Delta E + \frac{\kappa^2}{4M} \right)^2 / \left(\frac{\kappa^2 T}{M} \right) \right\} \sum_{\ell=|J'-J|}^{J'+J} C^2(J J' \ell; 00) |A_{n\ell}|^2 \\
 &\left. + a_1^2 \sum_{J'=1,3,5,\dots} (2J'+1) \exp \left\{ - \left(\epsilon + \Delta E + \frac{\kappa^2}{4M} \right)^2 / \left(\frac{\kappa^2 T}{M} \right) \right\} \sum_{\ell=|J'-J|}^{J'+J} C^2(J J' \ell; 00) |A_{n\ell}|^2 \right]
 \end{aligned}$$

where $\Delta E = E_J - E_{J'} + n\omega$, and for ortho-hydrogen

(29)

$$\begin{aligned}
 \left. \frac{d^2 \sigma_s}{d\Omega d\epsilon} \right|_{\text{ortho}} &= \frac{k}{3k_0} \sqrt{\frac{M}{\pi \kappa^2 T}} \sum_n \frac{1}{n!} \left(\frac{\kappa^2}{4M\omega} \right)^n \sum_{J=1,3,5,\dots} P_J \left[a_1^2 \sum_{J'=0,2,4,\dots} \right. \\
 &\cdot (2J'+1) \exp \left\{ - \left(\epsilon + \Delta E + \frac{\kappa^2}{4M} \right)^2 / \left(\frac{\kappa^2 T}{M} \right) \right\} \sum_{\ell=|J'-J|}^{J'+J} C^2(J J' \ell; 00) |A_{n\ell}|^2 \\
 &\left. + (3a_c^2 + 2a_1^2) \sum_{J'=1,3,5,\dots} (2J'+1) \exp \left\{ - \left(\epsilon + \Delta E + \frac{\kappa^2}{4M} \right)^2 / \left(\frac{\kappa^2 T}{M} \right) \right\} \sum_{\ell=|J'-J|}^{J'+J} C^2(J J' \ell; 00) |A_{n\ell}|^2 \right]
 \end{aligned}$$
(30)

Eqs. (29) and (30) have some special cases of interest. If the initial neutron energy is sufficiently less than 0.546 eV so that we satisfy the condition

$$\kappa^2/8M\omega \ll 1$$

for all scattering angles, then the vibrations need not be considered, and

$$A_{0\ell} = \int_{-1}^1 d\mu e^{\frac{i\kappa a \mu}{2}} P_{\ell}(\mu) = 2 i^{\ell} j_{\ell}\left(\frac{\kappa a}{2}\right),$$

where j_{ℓ} is the spherical Bessel function of order ℓ . Thus in this limit, Eqs. (29) and (30) become

$$\left. \frac{d^2 \sigma_s}{d\Omega d\epsilon} \right|_{\text{para}} = 4 \frac{k}{k_0} \sqrt{\frac{M}{\pi \kappa^2 T}} \sum_{J=0,2,4,\dots} P_J \left[a_c^2 \sum_{J'=0,2,4,\dots} (2J'+1) \exp \left\{ -(\epsilon + \Delta E + \frac{\kappa^2}{4M})^2 / (\frac{\kappa^2 T}{M}) \right\} \right. \\ \cdot \sum_{\ell=|J'-J|}^{J'+J} j_{\ell}^2 \left(\frac{\kappa a}{2} \right) C^2(J J' \ell; 00) + a_1^2 \sum_{J'=1,3,5,\dots} (2J'+1) \exp \left\{ -(\epsilon + \Delta E + \frac{\kappa^2}{4M})^2 / (\frac{\kappa^2 T}{M}) \right\} \\ \left. \cdot \sum_{\ell=|J'-J|}^{J'+J} j_{\ell}^2 \left(\frac{\kappa a}{2} \right) C^2(J J' \ell; 00) \right], \quad (31)$$

$$\left. \frac{d^2 \sigma_s}{d\Omega d\epsilon} \right|_{\text{ortho}} = \frac{4}{3} \frac{k}{k_0} \sqrt{\frac{M}{\pi \kappa^2 T}} \sum_{J=1,3,5,\dots} P_J \left[a_1^2 \sum_{J'=0,2,4,\dots} (2J'+1) \exp \left\{ -(\epsilon + \Delta E + \frac{\kappa^2}{4M})^2 / \left(\frac{\kappa^2 T}{M} \right) \right\} \right].$$

$$\cdot \sum_{l=|J'-J|}^{J+J'} j_l^2 \left(\frac{\kappa a}{2} \right) c^2(J J' l; 00) + (3a_c^2 + 2a_1^2) \sum_{J'=1,3,5,\dots} (2J'+1) \exp \left\{ -(\epsilon + \Delta E + \frac{\kappa^2}{4M})^2 / \left(\frac{\kappa^2 T}{M} \right) \right\}.$$

$$\cdot \sum_{l=|J-J'|}^{J+J'} j_l^2 \left(\frac{a}{2} \right) c^2(J J' l; 00) \quad (32)$$

Another special case is that of liquid hydrogen, where the molecules are in their ground rotational as well as vibrational states. Here we find:

$$\left(\frac{d^2 \sigma_s}{d\Omega d\epsilon} \right)_{\text{para}} = \frac{k}{k_0} \sqrt{\frac{M}{\pi \kappa^2 T}} \sum_n \frac{1}{n!} \left(\frac{\kappa^2}{4M\omega} \right)^n.$$

$$\left[a_c^2 \sum_{J'=0,2,4,\dots} (2J'+1) \exp \left\{ -(\epsilon + E_{J'} + n\omega - \frac{\kappa^2}{4M})^2 / \left(\frac{\kappa^2 T}{M} \right) \right\} \left| A_{n,J'} \right|^2 + \right. \quad (33)$$

$$\left. + a_1^2 \sum_{J'=1,3,5,\dots} (2J'+1) \exp \left\{ -(\epsilon + E_{J'} + n\omega - \frac{\kappa^2}{4M})^2 / \left(\frac{\kappa^2 T}{M} \right) \right\} \left| A_{n,J'} \right|^2 \right],$$

and for ortho-hydrogen,

$$\left(\frac{d^2 \sigma_s}{d\Omega d\epsilon} \right)_{\text{ortho}} = \frac{k}{3k_o} \sqrt{\frac{M}{\pi \kappa^2 T}} \sum_n \frac{1}{n!} \left(\frac{\kappa^2}{4M\omega} \right)^n. \quad (34)$$

$$\begin{aligned} & \left[a_1^2 \sum_{J'=0,2,4,\dots} \exp \left\{ -(\epsilon + E_{J'} - E_1 + n\omega - \frac{\kappa^2}{4M})^2 / (\frac{\kappa^2 T}{M}) \right\} \left((J'+1) \left| A_{n,J'+1} \right|^2 + J' \left| A_{n,J'-1} \right|^2 \right) + \right. \\ & \left. + (3a_c^2 + 2a_1^2) \sum_{J'=1,3,5,\dots} \exp \left\{ -(\epsilon + E_{J'} - E_1 + n\omega - \frac{\kappa^2}{4M})^2 / (\frac{\kappa^2 T}{M}) \right\} \left((J'+1) \left| A_{n,J'+1} \right|^2 + J' \left| A_{n,J'-1} \right|^2 \right) \right] \end{aligned}$$

Actually no specifically liquid effects have been included in Eqs. (33) and (34), but as indicated earlier, these are expected to be of importance only for extremely low energy neutrons ($E_0 < .002$ ev).

The integrals A_{nl} can all be evaluated by recursion from the single integral A_{00} , and these recursion relations are given in Appendix C.

At room temperature one might ask if the inclusion of spin correlations is important. The answer seems to be affirmative, since, as an investigation of P_J readily shows, at this temperature only the rotational states $J=0,1,2,3$ are present in any appreciable amount.

The cross-section for scattering from an ortho-para mixture is given by

$$\frac{d^2 \sigma_s}{d\Omega d\epsilon} = \frac{N_{\text{para}}}{N_{\text{para}} + N_{\text{ortho}}} \left. \frac{d^2 \sigma_s}{d\Omega d\epsilon} \right|_{\text{para}} + \frac{N_{\text{ortho}}}{N_{\text{para}} + N_{\text{ortho}}} \left. \frac{d^2 \sigma_s}{d\Omega d\epsilon} \right|_{\text{ortho}},$$

where N_{para} and N_{ortho} are the number densities of para and ortho molecules respectively. For $kT \gg .015$, $N_{\text{para}}/N_{\text{ortho}} = 1/3$, and at room temperature ($.0258 \text{ ev}$), $N_{\text{para}}/N_{\text{ortho}} = 1/2.91$.

IV. INTERFERENCE EFFECTS

So far our calculations have been for the "self" scattering, i.e., referring to a single H_2 molecule; and for gaseous hydrogen the influence of the interference of neutron waves scattered from different molecules will be very small. For liquid hydrogen however, interference effects might be of importance for very low neutron energies where para \rightarrow para transitions are the only competition, i.e., for neutron energies below .015 volts. Even for these low energies, the para \rightarrow para cross-section will dominate. Interference effects for liquid hydrogen in terms of the translational correlation function for neutron energies below the first vibrational level were calculated by Sarma⁽⁴⁾, but for completeness we quote the formula here with the inclusion of the vibrations:

$$\frac{d^2\sigma_{int}}{d\Omega d\epsilon} = \frac{a_c^2}{2\pi} \frac{k}{k_0} \int_{-\infty}^{\infty} dt e^{+i\epsilon t} \sum_{l \neq j} \langle \psi_{it} | e^{-i\vec{\kappa} \cdot \vec{r}_j(0)} e^{+i\vec{\kappa} \cdot \vec{r}_l(t)} | \psi_{it} \rangle_T \quad (34)$$

$$\left[\int_{-1}^1 d\mu \cos\left(\frac{\kappa a \mu}{2}\right) e^{-\frac{\kappa^2 \mu^2}{8M\omega}} \right]^2 .$$

For a free gas the translational correlation function

$$\sum_{l \neq j} \langle \psi_{it} | e^{-i\vec{\kappa} \cdot \vec{r}_j(0)} e^{+i\vec{\kappa} \cdot \vec{r}_l(t)} | \psi_{it} \rangle_T$$

vanishes.

V. NUMERICAL RESULTS AND DISCUSSION

Equations (29) and (30) have been programmed for computation on the IBM 7044. In particular, Figure (1) shows the total cross-section (per atom) for scattering from both ortho- and para-hydrogen gas at 20.4°K for initial neutron energies up to 1 ev. Notice that the para-hydrogen cross-section rises rapidly as E_0 approaches .022 ev, which is just the energy the neutron needs in the laboratory frame to cause the rotational transition $J = 0 \rightarrow J = 1$, from para- to ortho-hydrogen. Also shown in Figure (1) are some experimental points by G. Squires and A. Stewart⁽⁷⁾. In Figures (2) through (5) we show the computed double differential cross section $\frac{d^2\sigma}{dE d\Omega}$ for two different initial energies and scattering angles. For $E_0 = 1.0$ ev and $\theta = 32^{\circ}$ one can clearly recognize the structure due to the rotational transitions superimposed on the zero and one phonon contributions to the cross section. The structure is more pronounced for para- than for ortho-hydrogen, since in the case of para-hydrogen only the odd rotational levels make a significant contribution, whereas all levels contribute to the ortho cross-section. The even levels in ortho-hydrogen contribute roughly twice as much as the odd levels as seen in Eq. (30). At a scattering angle of 55° the structure due to the rotational transitions is still present but appears somewhat smoothed out because of the larger recoil energy. The same comments apply to the curves corresponding to $E_0 = 0.22$, although here only the zero phonon term can contribute to the cross-sections.

In summary then, the results given in this report predict the scattering cross-sections from H_2 and D_2 for incident neutron energies up to roughly 3 ev and all practical temperatures. For neutron energies above a few volts the calculated total cross-sections for both ortho- and para-hydrogen

tend to the free atom limit as they should, and for low neutron energies, so that only the first rotational transition occurs, the results are identical to those of Sarma.⁽⁴⁾ The calculated total cross-section agrees reasonably well with the available experimental data, and as the experimental errors decrease with increasing neutron energy so does the agreement between theory and experiment improve. Since the differential cross-sections are far more sensitive to the details of the model than the total cross-section, it is hoped that differential experiments planned and in progress⁽⁸⁾ for various ranges of neutron energy will allow a rigorous test of the theory.

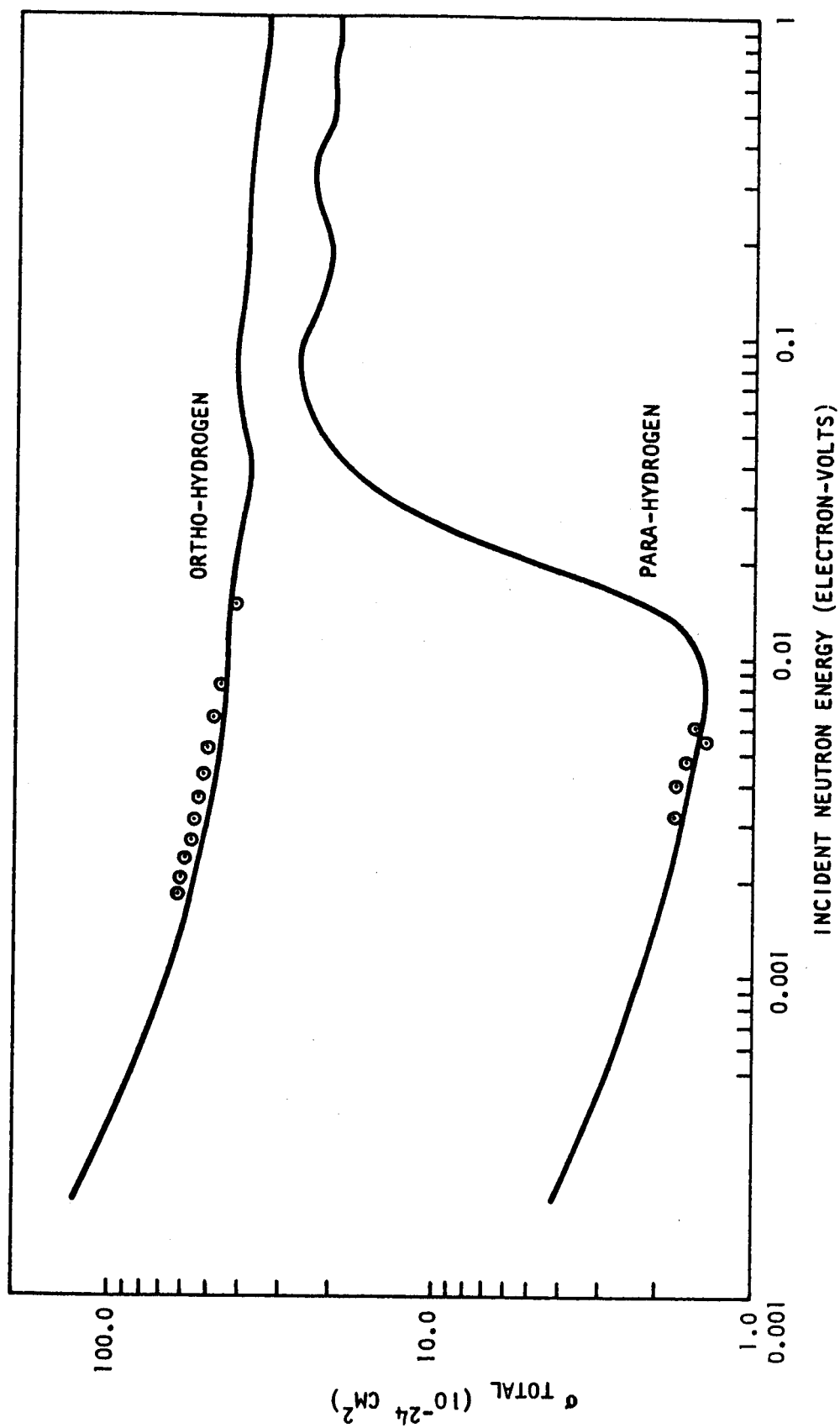


Fig. 1 - The total cross-section for both para- and ortho-hydrogen at a temperature $T = 20.4^\circ \text{K}$.

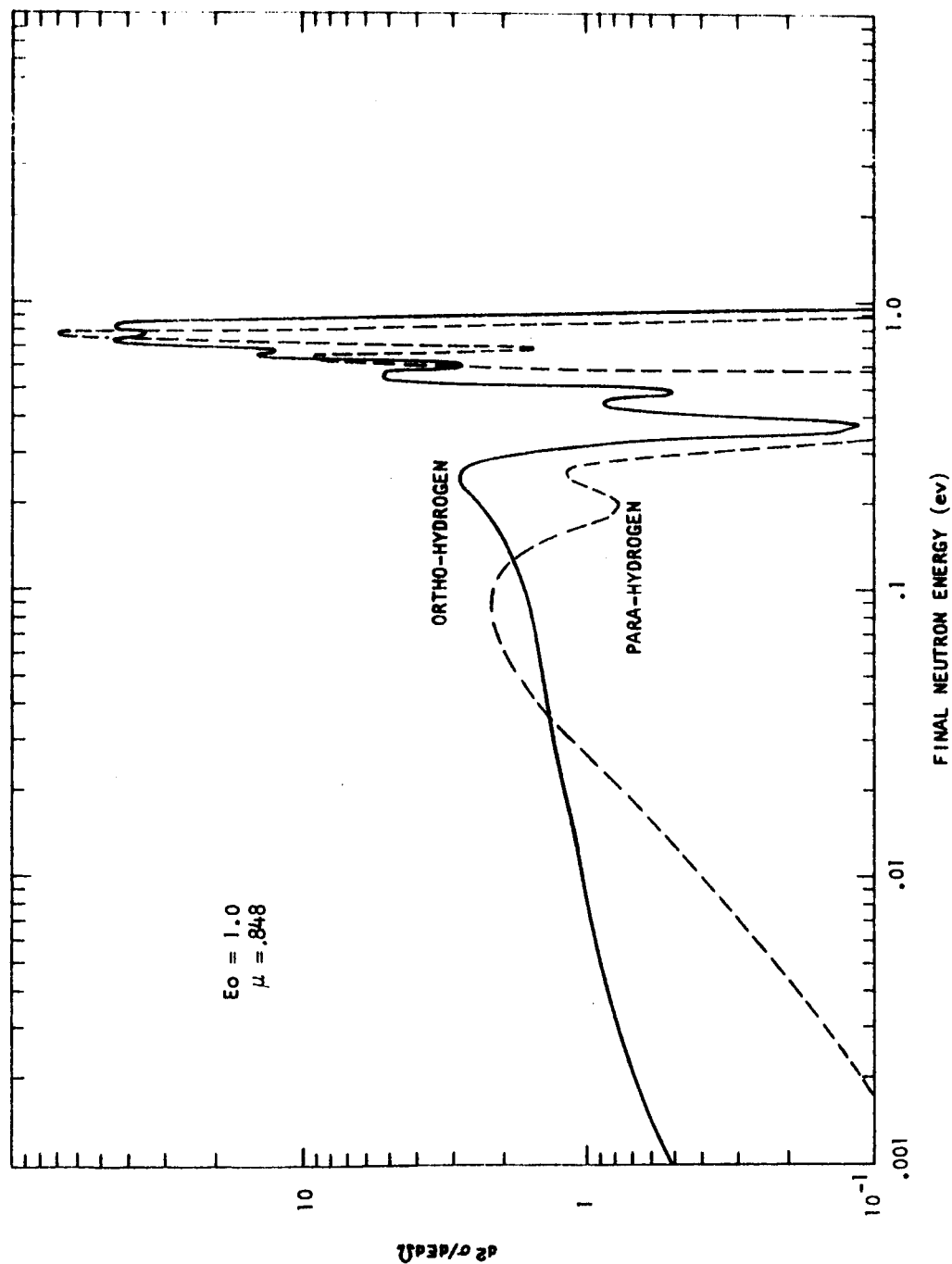


Fig. 2 - The double differential cross-section $\frac{d^2\sigma}{dE_d\Omega}$ for an initial neutron energy of 1.0 ev and a scattering angle of 32° , for both para- and ortho-hydrogen at $T = 20.40^\circ\text{K}$.

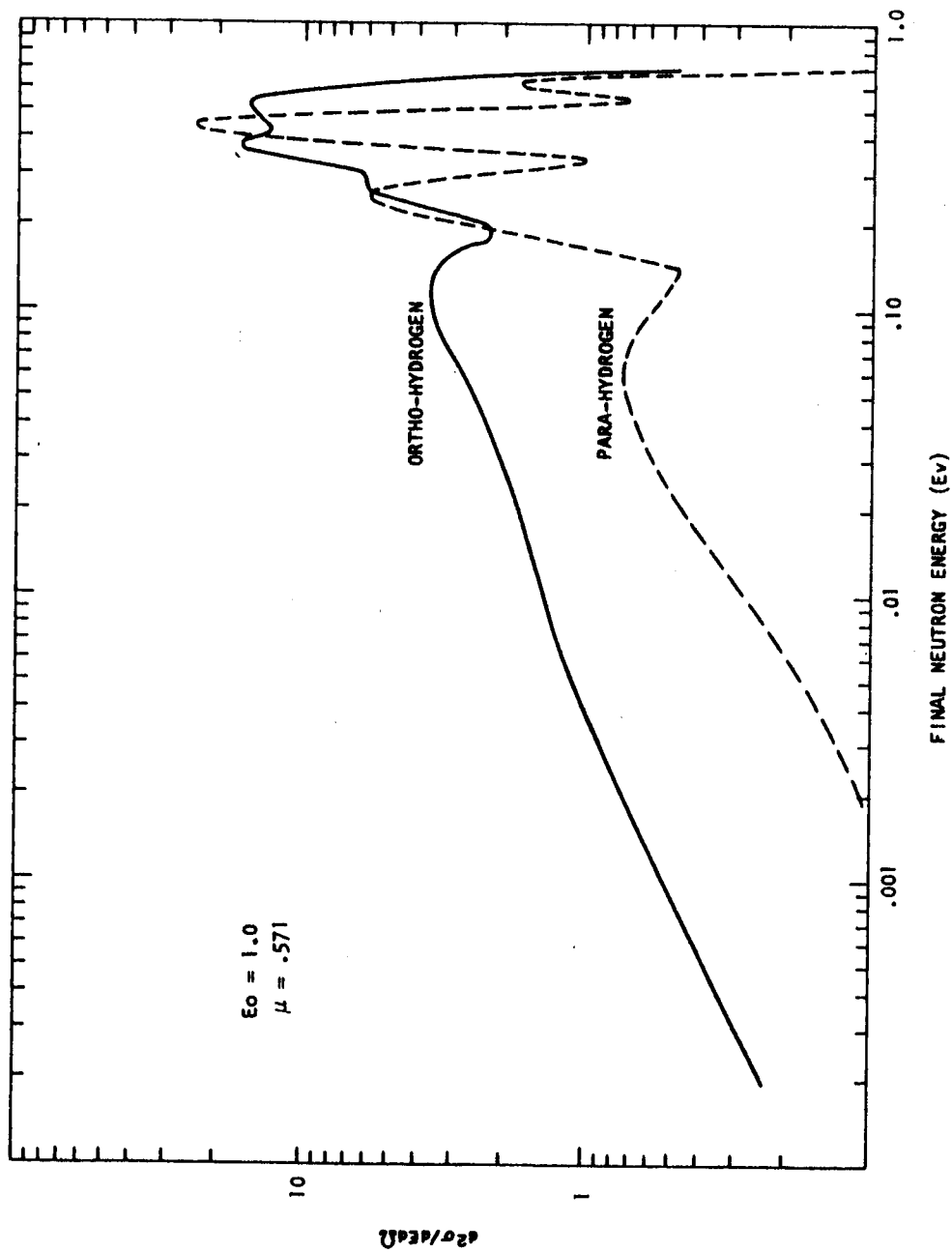


Fig. 3 - The double differential cross-section $d^2\sigma/dE d\Omega$ for an initial neutron energy of 1.0 eV and a scattering angle of 55° , for both para- and ortho-hydrogen at $T = 20.4^\circ\text{K}$.

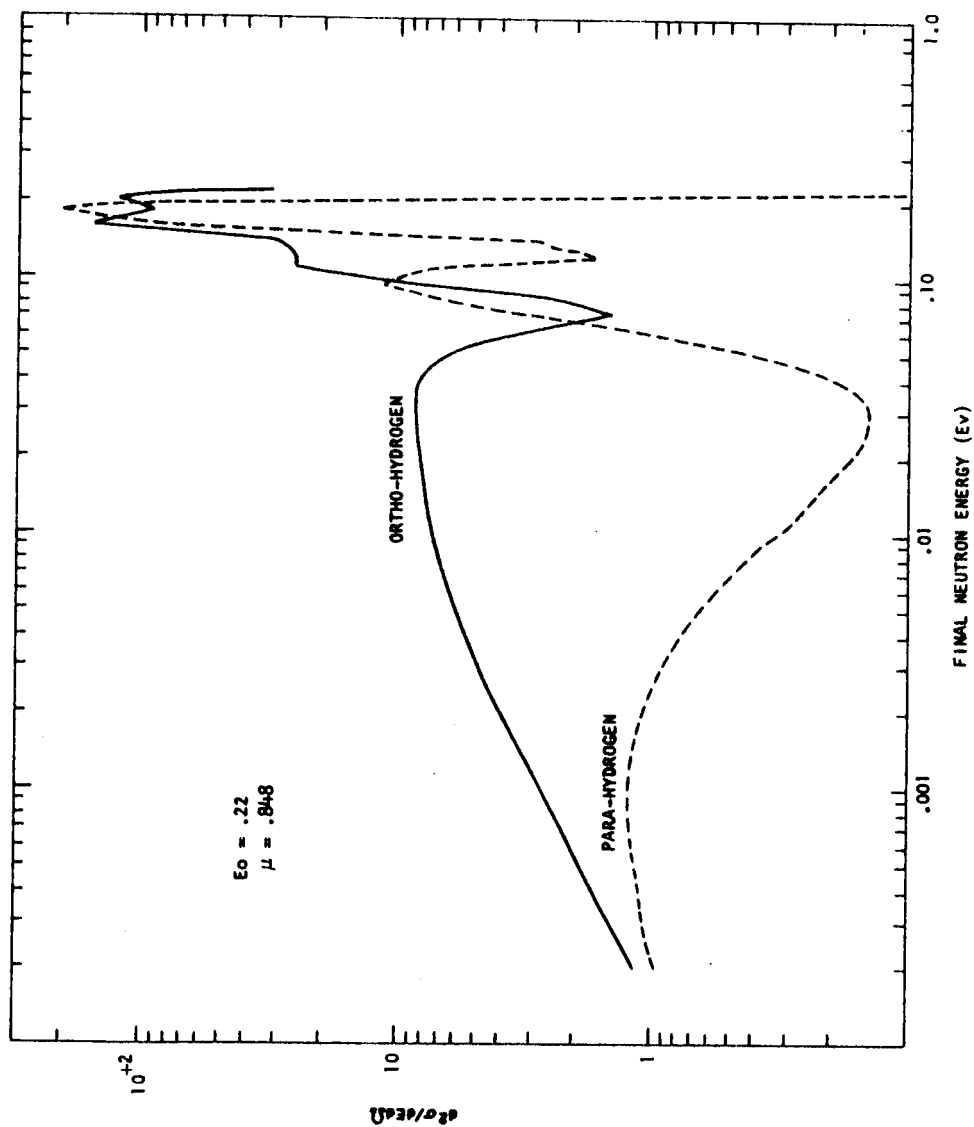


Fig. 4 - The double differential cross-section $\frac{d^2\sigma}{dE_d\Omega}$ for an initial neutron energy of .22 eV and a scattering angle of 32° , for both para- and ortho-hydrogen at $T = 20.4^\circ\text{K}$.

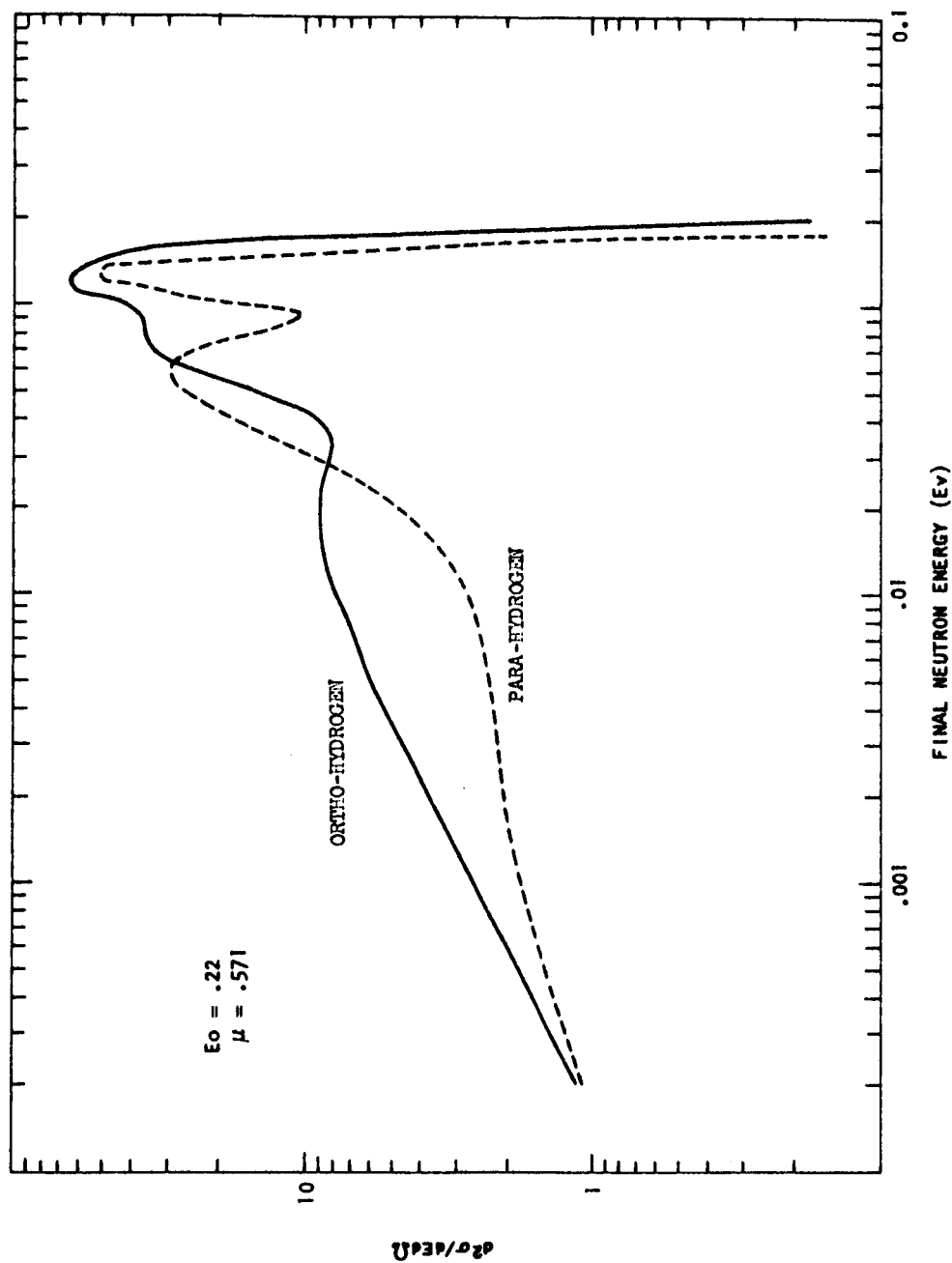


Fig. 5 - The double differential cross-section $d^2\sigma/dE d\Omega$ for an initial neutron energy of .22 eV and a scattering angle of 55° , for both para- and ortho-hydrogen at $T = 20.4^\circ\text{K}$.

APPENDIX A

D₂ Molecule

Aside from the trivial difference in masses, the only difference between the H₂ and the D₂ molecule lies in the spin correlation. In this appendix we calculate the cross-sections for the scattering of neutrons from D₂ by evaluating the effect of the spin correlations, and using the H₂ results already obtained for the vibrations, rotations and translations.

For D₂ we find (with the same notation as in section II)

$$\begin{aligned}
 A &= a_1 e^{\frac{i\vec{\kappa} \cdot \vec{R}}{2}} + a_2 e^{\frac{-i\vec{\kappa} \cdot \vec{R}}{2}} \\
 &= P \cos \left(\frac{\vec{\kappa} \cdot \vec{R}}{2} \right) + i Q \sin \left(\frac{\vec{\kappa} \cdot \vec{R}}{2} \right), \quad (A-1)
 \end{aligned}$$

where

$$P = (2a_{\text{coh}} + \sqrt{2} a_{\text{inc}} \vec{S} \cdot \vec{\sigma}), \quad (A-2)$$

$$Q = \sqrt{2} a_{\text{inc}} \vec{\sigma} \cdot (\vec{I}_1 - \vec{I}_2) \quad (A-3)$$

and $\vec{S} = \vec{I}_1 + \vec{I}_2$ is the total spin of the molecule. Since the deuteron has spin one, the symmetric ("ortho") nuclear spin eigenfunctions of the D₂ molecule are those of spin 0 and 2, and these correspond to even values of J. The antisymmetric ("para") states have spin 1 and odd J values. It can readily be shown that the total spin operator \vec{S} has nonzero matrix elements only between states of the same total $\vec{S}(\vec{S}^2 = S(S+1))$ and that the operator Q

has nonzero matrix elements only between states of total S differing by 1.

It follows from this, and also from the conservation of spin angular momentum ($\Delta S = \pm 1$), that the transitions $S = 0 \longleftrightarrow S = 2$ cannot occur.

Using the Eqs. (11), (12), (14), (A-1) we find in this case

$$g(t)_{S=S'} = \frac{1}{2} \left[8a_{\text{coh}}^2 + S(S+1)a_{\text{inc}}^2 \right] \sum_J \frac{P_{JS}}{2J+1} \sum_{J'} e^{i(E_{J'} - E_J)t} \sum_{n=0}^{\infty} e^{in\omega t} \cdot$$

$$\cdot \sum_{J_z' J_z} \left| \langle J' J_z', n | \cos \left(\frac{\vec{\kappa} \cdot \vec{R}}{2} \right) | J J_z, n=0 \rangle \right|^2,$$

where $\sum_{J'}$ implies that J' and J have the same parity, and for J' , J both even, S is 0 or 2, while for J' , J both odd S is 1.

Similarly, Eqs. (17), (19) and (A-3) result in

$$g(t)_{S \neq S'} = \frac{a_{\text{inc}}^2}{2} (8 - S(S+1)) \sum_J \frac{P_{JS}}{2J+1} \sum_{J'}'' e^{i(E_{J'} - E_J)t} \sum_n e^{in\omega t} \cdot$$

$$\cdot \sum_{J_z J_z'} \left| \langle J' J_z', n | \sin \left(\frac{\vec{\kappa} \cdot \vec{R}}{2} \right) | J J_z, n=0 \rangle \right|^2,$$

where $\sum_{J'}''$ implies that J' and J have opposite parity, and for J even, S is 0 or 2, while for J odd, S is 1.

The above results now give, in complete analogy with the way Eqs. (29) and (30) were obtained, the "self" cross-section for ortho-deuterium:

$$\begin{aligned} \left. \frac{d^2 \sigma_s}{d\Omega d\epsilon} \right|_{\text{ortho}} &= \frac{k}{k_0} \sqrt{\frac{M_d}{\pi \kappa^2 T}} \sum_n \frac{1}{n!} \left(\frac{\kappa^2}{4M_d \omega} \right)^n \sum_{J=0,2,4,\dots} P_J \left[(a_{\text{coh}}^2 + \frac{5}{8} a_{\text{inc}}^2) \cdot \right. \\ &\cdot \sum_{J'=0,2,4,\dots} (2J'+1) \exp \left\{ -(\epsilon + \Delta E + \frac{\kappa^2}{4M_d})^2 / \left(\frac{\kappa^2 T}{M_d} \right) \right\} \sum_{\ell=|J'-J|}^{J'+J} C^2(J J' \ell; 00) |A_{n\ell}|^2 \\ &+ \frac{3}{8} a_{\text{inc}}^2 \sum_{J'=1,3,5,\dots} (2J'+1) \exp \left\{ -(\epsilon + \Delta E + \frac{\kappa^2}{4M_d})^2 / \left(\frac{\kappa^2 T}{M_d} \right) \right\} \sum_{\ell=|J'-J|}^{J'+J} C^2(J J' \ell; 00) |A_{n\ell}|^2 \left. \right] \end{aligned}$$

(A-4)

and for para-deuterium we find:

$$\begin{aligned} \left. \frac{d^2 \sigma_s}{d\Omega d\epsilon} \right|_{\text{para}} &= \frac{k}{4k_0} \sqrt{\frac{M_d}{\pi \kappa^2 T}} \sum_n \frac{1}{n!} \left(\frac{\kappa^2}{4M_d \omega} \right)^n \sum_{J=1,3,5,\dots} P_J \left[3a_{\text{inc}}^2 \sum_{J'=0,2,4,\dots} (2J'+1) \cdot \right. \\ &\cdot \exp \left\{ -(\epsilon + \Delta E + \frac{\kappa^2}{4M_d})^2 / \left(\frac{\kappa^2 T}{M_d} \right) \right\} \cdot \sum_{\ell=|J'-J|}^{J'+J} C^2(J J' \ell; 00) |A_{n\ell}|^2 \\ &+ (4a_{\text{coh}}^2 + a_{\text{inc}}^2) \sum_{J'=1,3,5,\dots} (2J'+1) \exp \left\{ -(\epsilon + \Delta E + \frac{\kappa^2}{4M_d})^2 / \left(\frac{\kappa^2 T}{M_d} \right) \right\} \sum_{\ell=|J'-J|}^{J'+J} C^2(J J' \ell; 00) |A_{n\ell}|^2 \left. \right] \end{aligned}$$

(A-5)

where M_d is the deuteron mass. The energy levels for D_2 are given by:

$$E_J = \frac{\hbar^2 J(J+1)}{4M_d a^2} = .0071 \frac{J(J+1)}{2} \text{ ev.}$$

and thus for low temperatures ($\lesssim .02$ ev) spin correlations will be important for deuterium.

APPENDIX B

Here the values of the Clebsch-Gordon coefficients $C^2(J J' \ell; 00)$ are given for $J = 0, 1, 2, 3$. The analytic expression for $C(J J' \ell; 00)$ is given in reference (6). For simplicity in notation we define the quantity $(J J' | \ell)$ so that

$$(J J' | \ell) = (2J' + 1) C^2(J J' \ell; 00).$$

The results are:

$$J = 0$$

$$(0 J' | \ell) = 2J' + 1,$$

$$J = 1$$

$$(1 J' | J' + 1) = J' + 1, \quad (1 J' | J' - 1) = J'$$

$$J = 2$$

$$(2 J' | J' + 2) = \frac{3}{2} \frac{(J' + 2)(J' + 1)}{2J' + 3}, \quad (2 J' | J') = \frac{J'(J' + 1)(2J' + 1)}{(2J' + 3)(2J' - 1)},$$

$$(2 J' | J' - 2) = \frac{3}{2} \frac{J'(J' - 1)}{2J' - 1}$$

$$J = 3$$

$$(3 J' | J' + 3) = \frac{5}{2} \frac{(J' + 3)(J' + 2)(J' + 1)}{(2J' + 5)(2J' + 3)}, \quad (3 J' | J' + 1) = \frac{3}{2} \frac{(J' + 2)(J' + 1)J'}{(2J' + 5)(2J' - 1)}$$

$$(3 J' | J' - 1) = \frac{3}{2} \frac{(J' + 1)J'(J' - 1)}{(2J' + 3)(2J' - 3)}, \quad (3 J' | J' - 3) = \frac{5}{2} \frac{J'(J' - 1)(J' - 2)}{(2J' - 1)(2J' - 3)}$$

$$J = 4$$

$$(4 J' | J' + 4) = \frac{35}{8} \frac{(J' + 4)(J' + 3)(J' + 2)(J' + 1)}{(2J' + 7)(2J' + 5)(2J' + 3)},$$

$$(4 J' | J' + 2) = \frac{5}{2} \frac{(J' + 3)(J' + 2)(J' + 1)J'}{(2J' + 7)(2J' + 3)(2J' - 1)},$$

$$(4 J' | J') = \frac{9}{4} \frac{(2J' + 1)(J' + 2)(J' + 1)(J' - 1)J'}{(2J' + 5)(2J' + 3)(2J' - 1)(2J' - 3)},$$

$$(4 J' | J' - 2) = \frac{5}{2} \frac{(J' + 1)J'(J' - 1)(J' - 2)}{(2J' + 3)(2J' - 1)(2J' - 5)},$$

$$(4 J' | J' - 4) = \frac{35}{8} \frac{J'(J' - 1)(J' - 2)(J' - 3)}{(2J' - 1)(2J' - 3)(2J' - 5)}$$

APPENDIX C

Recursion relations useful in evaluating the $A_{n,\ell}$, which are defined by Eq. (27), are presented here. First the recursion relations of interest here for the $P_\ell(\mu)$ are:

$$(2\ell+1)\mu P_\ell(\mu) = (\ell+1) P_{\ell+1}(\mu) + \ell P_{\ell-1}(\mu), \quad (\text{A-1})$$

$$(2\ell+1) P'_\ell(\mu) = P'_{\ell+1}(\mu) - P'_{\ell-1}(\mu). \quad (\text{A-2})$$

We put a superscript (+) on $C_{n,\ell}$ if ℓ is even, and a (-) for odd ℓ . Therefore

$$C_{0,\ell}^{(+)} = \int_{-1}^1 d\mu P_\ell(\mu) \cos(\alpha\mu) e^{-\lambda\mu^2} = A_{0,\ell}^{(+)} \quad (\text{A-3})$$

$$C_{0,\ell}^{(-)} = \int_{-1}^1 d\mu P_\ell(\mu) \sin(\alpha\mu) e^{-\lambda\mu^2} = -iA_{0,\ell}^{(-)} \quad (\text{A-4})$$

where $\alpha = \kappa a/2$, $\lambda = \kappa^2/8M\omega$. By using (A-1) and (A-2):

$$C_{0,1}^{(-)} = - (1/\lambda) e^{-\lambda} \sin \alpha + (\alpha/2\lambda) C_{0,0}^{(+)}$$

$$C_{0,2}^{(+)} = - (3/2\lambda) e^{-\lambda} \cos \alpha + ((3/2\lambda)-1) C_{0,0}^{(+)} - (3\alpha/4\lambda) C_{0,1}^{(-)},$$

$$\begin{aligned}
c_{0,l+2}^{(+)} &= \frac{2l+1}{2\lambda(l+2)(2l-1)} \left[(2l-1)(2l+3)+2\lambda \right] c_{0,l}^{(+)} + \frac{(2l+3)}{2\lambda(l+2)} \alpha (c_{0,l-1}^{(-)} - c_{0,l+1}^{(-)}) \\
&\quad + \frac{(2l+3)(l-1)}{(l+2)(2l-1)} c_{0,l-2}^{(+)} , \quad l = 2, 4, \dots
\end{aligned} \tag{A-5}$$

$$\begin{aligned}
c_{0,l+2}^{(-)} &= \frac{2l+1}{2\lambda(l+2)(2l-1)} \left[(2l-1)(2l+3)+2\lambda \right] c_{0,l}^{(-)} - \frac{2l+3}{2\lambda(l+2)} \alpha (c_{0,l-1}^{(+)} - c_{0,l+1}^{(+)}) \\
&\quad + \frac{(2l+3)(l-1)}{(l+2)(2l-1)} c_{0,l-2}^{(-)} , \quad l = 1, 3, 5, \dots
\end{aligned}$$

The recursion relation for $n \neq 0$ is given simply by

$$c_{n,l}^{(\pm)} = \frac{l+1}{2l+1} c_{n-1,l+1}^{(\mp)} + \frac{l}{2l+1} c_{n-1,l-1}^{(\mp)} .$$

We see then that the only integral that need be computed directly is $c_{0,0}^{(+)}$, all the rest following from recursion relations.

LIST OF SYMBOLS

ψ_i	initial wave function
ψ_f	final wave function
k_o	initial neutron momentum
k	final neutron momentum
e	neutron energy transfer
\vec{k}	neutron momentum transfer
E_i	initial neutron energy
E_f	final neutron energy
\vec{r}_m	position vector of m-th atom
a_m	bound scattering length of m-th atom
\vec{R}	relative coordinate of two atoms in same molecule
\vec{r}_l	position coordinate of molecular centre-of-mass
σ	cross-section
Ω	solid angle
\vec{S}	molecular spin
J	orbital angular momentum of molecule
$\vec{\sigma}$	neutron spin vector
P_{Js}	statistical weight of state J with spin S.
w	quantum of vibrational energy
n	vibrational quantum number
E_J	rotational energy of state J
a_{coh}	coherent scattering amplitude
a_{inc}	incoherent scattering amplitude
\vec{I}_m	nuclear spin
a_+	triplet scattering length
a_-	singlet scattering length

\vec{I}	total nuclear spin
M	mass of hydrogen atom
b	annihilation operator for vibrations
b^+	creation operator for vibrations
Θ	angle between vectors \vec{k} and \vec{r}
μ	$\cos \Theta$
$Y(\Theta, \phi)$ $J \ J_z$	spherical harmonic
a	equilibrium separation between atoms in H_2 molecule
$C(l, l_2 l; m, m_2)$	Clebsch-Gordon coefficient
δ_{J_z, J_z}	Kronecker-delta
$P_\ell(u)$	Legendre polynomial of order ℓ
T	temperature
ΔE	$E_{J'} - E_J + n\omega$
j_ℓ	spherical Bessel function of order ℓ
M_d	mass of deuterium atom.

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